Study of photorefractive beam coupling in $Sn_2P_2S_6$ in reflection grating geometry

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Executive summary

The project belongs to the technology Area "Physics: Optics and Lasers" and it deals with the development of coherent optical amplifiers based on selfdiffraction from the dynamic gratings recorded by the weak signal beam to be amplified and much more powerful coherent pump beam. The purpose of this project was to find the conditions under which a relatively new crystal with rather fast nonlinear response, tin hypothiodiphosphate ($Sn_2P_2S_6$), can ensure the enhanced beam coupling over other currently used photorefractive materials.

To achieve this goal we intended to grow $Sn_2P_2S_6$ deliberately doped with special impurity and to optimize the grating recording geometries in a way that allow for taking profit from optimum combination of crystal electrooptic constants and dielectric constants. We expected to improve at least two times the beam coupling gain factor in reflection geometry for tin hypothiodiphosphate and to develop recommendations for further improvement of this principal parameter of optical amplifiers and optical limiters.

Two principal ways to make larger the two-beam-coupling gain factor were investigated, one technological, via deliberate doping of crystal during growing procedure and the other one based on optimization of interaction geometry and on appropriate choice of polarization of the interacting light waves. Because of incomplete available information about defect center content, material constants, and electrooptic and nonlinear properties of $Sn_2P_2S_6$ both these approaches were new and necessary. Both ways brought positive results that contributed to achievement of principal goal to reach two-times larger value of gain factor for contradirectional beam coupling.

Different impurities that increase the absorption in red and near infraded domain of spectrum and ensure pronounced photoconductivity were tested, Fe, Sn, Pb, Te, Sb. Most of samples were already grown before, within the programs funded by other projects; they were tested now for the particular objective of running project. The best results at present were achieved with Te doped material, for which an obvious increase of the effective trap density was found. This pushed us to study this dopant more carefully and to grow samples with increasing content of Te. It was found that the effective trap density reaches its saturation for roughly 3 wt.% of Te in initial components. At the same time, the increasing Te contents leads to undesirable rise of the absorption. The optimum Te content was found to be about 1 wt.%; for this concentration the gain in coupling is still larger than the loss which is due to increasing lineart absorption. Our preliminary experiments with Sb-doped gave us an optimism that this dopant could be even better alternative to Te especially in the near infrared.

To improve coupling via optimization of the interaction geometry and polarization it was necessary at first to evaluate missing Pockels tensor components. We did it by using both a traditional technique, by measuring phase retardation versus applied electric field and also with photorefractive technique, by measuring orientation dependences and polarization dependences of two-beam coupling gain. The fit of dependences calculated for known structure of the Pockels tensor for *m*-symmetry crystal to the measured dependences gave quite reliable data for the ratios of different Pockels tensor components. With the known values for few diagonal components this procedure allows for evaluation of many other components. With the exception of two components, r_{322} and r_{122}, we have now nearly full description of linear electrooptic effect in $Sn_2P_2S_6$. When working in this direction we detected, for the

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14. ABSTRACT
This report results from a contract tasking National Academy of Sciences as follows: This project belongs to the technology Area Physics: Optics and Lasers and more precisely it deals with the development of coherent optical amplifiers based on selfdiffraction from the dynamic gratings recorded by the weak signal beam to be amplified and much more powerful coherent pump beam. The main strategic goal of this project consists, in fact, not in amplification of the signal but in deamplification (depletion) of the pump wave, as much effective as possible. To make a step in achieving this goal, a relatively new photorefractive material will be grown, characterized and tested, Tin Hypothiodiphosphate (Sn2P2S6) in the Institute of Solid State Physics and Chemistry, Uzhgorod and Institute of Physics, Kiev. The first institute possesses facilities necessary to grow these crystals, to put them into single domain state and to cut and polish samples while the second institute has a long lasting expertise in studies of nonlinear wave mixing in photorefractive materials.
The purpose of this project is to find the conditions under which tin hypothiodiphosphate (Sn2P2S6) can ensure the enhanced beam coupling over other currently used materials.
To do it new, deliberately doped Sn2P2S6 will be grown and grating recording will be done in new interaction geometries to take profit from optimum combination of crystal electrooptic constants and dielectric constants.
We expect to improve at least two times the beam coupling gain factor in reflection geometry known today for tin hypothiodiphosphate and to develop recommendations for further improvement of this principal parameter of optical amplifiers and optical limiters.

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first time for $Sn_2P_2S_6$, the anisotropic selfdiffraction (appearance of additional diffracted spot with the polarization orthogonal to that of the recording waves) and anisotropic diffraction from isotropically recorded grating. This is a direct prove of nonvanishing nondiagonal Pockels coefficients, r_{131} and r_{313}. Unfortunately, any of new evaluated Pockels coefficients is larger or even comparable to the largest known before, r_{111} = 174 pm/V.

With the detailed information on electrooptic properties and with Te-doped samples that feature the increased trap density we started optimizing the geometry of conradirectional beam coupling and finally get more than two-times improvement as compared to nominally undoped crystals in traditional orientation of grating vector along the crystallographic axis. Two beam coupling strength more than 2,4 has been achieved for specially tilted $Sn_2P_2S_6$:Te sample. We believe, this value can be increased further for specially cut samples.

We analyzed also the other possible ways of improvement of the two-beam coupling and proposed a technique of considerable improvement of the transient gain for the crystals possessing two types of movable charge carriers with considerably different dielectric relaxation times. The technique proposed consists in periodic modulation of the phase difference of two interacting waves between two discret states, zero and PI. The improvement of gain is achieved if modulation frequency is smaller than the decay rates for both types of movable charge carriers; it takes place only in condition of space charge limitations (i.e., just for the case of contradirectional beam coupling in $Sn_2P_2S_6$.

Cooperation with foreign collaborators

All quarterly reports on present Project have always been submitted for approval of our collaborator Dr. Dean Evans from US Air Force Research Laboratory in Dayton. From time to time the description of current results and/or measured dependences have been sent via electronic mail to Dr. D. Evans or Dr. Gary Cook. Especially active this correspondence was just before the Photorefractive Workshop organized by US AFRL in St.Petersburg, Florida, in August 2006. A joint contribution "Optimizing beam coupling in $Sn_2P_2S_6$ " (*S. Odoulov, A. Shumelyuk, D. Evans and G. Cook)* was prepared and presented during the Workshop. Very intensive e-mail correspondence we had also in the beginning of this year when finalizing common publication submitted (and now accepted) to Optics Letters.

Publications

A part of the Project results is described in two articles accepted for publication:

1. ***Transient gain enhancement in photorefractive crystals with two types of movable charge carriers*** (A. Shumelyuk, A. Hrygorashchuk, S. Odoulov, and D. Evans) **Optics Letters,** vol.**32**, # 14, Jul.15, 2007.
2. ***Spectral sensitivity of nominally undoped photorefractive***
(A. Shumelyuk, S. Odoulov, O. Oleinik, G. Brost and A. Grabar), Applied Physics B – Lasers and Optics, 2007

Prospects of future development.

How the results of this Project will be used depends solely on decision of our US partners from the US AFRL. Our task to get at least two-times improvement of the beam coupling gain factor is accomplished in full. It should be added however that when working on this project we got more clear vision how the gain factor in $Sn_2P_2S_6$ can be improved further, both via technological means and via modification of interaction geometry. At present we apply for a new Partner Project (STCU/EOARD) that is called <u>New ways of beam coupling enhancement in low-symmetry photorefractive crystals.</u>

Project manager

Serguey ODOULOV

For the Coordinating Institution

Approved

Dean EVANS
<enter position of authorized representative
of the coordinating institution>

Project main idea

The beam-coupling gain that we need to improve depends in any photorefractive crystal on **effective electrooptic constant** and on **light-induced space charge field.** The components of electrooptic tensor (Pockels tensor) are constants of the material; they depend on temperature and wavelength but usually they cannot be strongly affected by adding impurities or by changing the crystal growth technology. For relatively new nonlinear crystal $Sn_2P_2S_6$ however the data on Pockels tensor are incomplete and the scatter of data reported for few measured components is too big. One could expect, therefore, to find among the unknown Pockels tensor components those with equal or even larger absolute values as that already estimated. This could suggest the particular crystal cuts where an effective electrooptic constant might become larger than that known for sample cut along the crystallographic axes.

The space charge field induced inside the sample, on the contrary, is strongly affected by the growth procedure. It can be much smaller than the ultimate value that is imposed by the driving electric field (diffusion field, applied external field, or photovoltaic field) because of insufficient density of empty traps that can store the photoexcited and redistributed charges. To large extent the effective trap density can be affected by growing the crystal with deliberately added impurities or by aftergrowth treatment that can introduce the desirable defects.

Technical approach

Two general approaches were used in this Project, **one purely technological,** aiming in reduction of the space charge limitation via increasing of the effective trap density, and **the other one purely optical,** based on finding the best orientation and polarization of interacting light waves, that profit from largest effective electrooptic tensor component.

Technical progress overview

<u>The workplan of the Project included four large tasks</u>:

- Growth, preparation and optical characterization of new nominally undoped $Sn_2P_2S_6$ crystals.
- Photorefractive characterization of the available and new grown $Sn_2P_2S_6$ samples.
- Experimental evaluation of nondiagonal components of the Pockels tensor
- Growth, preparation and optical characterization of new $Sn_2P_2S_6$ crystals with deliberately introduced impurity.

During one year **8** $Sn_2P_2S_6$ samples have been grown, cut, oriented along crystallographic axes and optically finished in Uzhgorod. For most of them the optical quality test was done, the absorption spectra and photoconductivity were measured.
2 nominally undoped and **2** deliberately doped with Te samples have been delivered to Kiev group for detailed study of photorefractive properties.

With the combined efforts (technological and optical) the main goal of the Project has been achieved, the beam-coupling gain factor about 2 cm^{-1} for counterpropagating light beams was measured, which is more than two times larger that the best value (0.6-0.8) cm^{-1}, known before this work was started [*M. Weber, G. von Bally, A. Shumelyuk, and S. Odoulov, Reflection-type photorefractive gratings in Tin Hypotiodiphosphate, Appl. Phys. B, 74, #1, pp. 29-33, 2002*].

Summary of personnel commitment.

The tasks break between researches involved in present Project was roughly as it is listed below. It was not very strict however and, except the main crystal grower Dr Stoyka, every other participant was working quite often with one or two colleagues solving the experimental task or fitting the data to calculated dependences.

Dr. Ivan Stoyka concentrated uniquely on the cleaning the initial components and crystal synthesis;

Dr. Yulian Vysochansky measured the optical and photoelectric spectra, as well as dielectric parameters. He also analyzed the obtained results comparing them with that available from literature.

Dr. Alexander Grabar performed the sample fabrication (orientation, cutting, polishing and poling), measurements of the photorefractive parameters (two-wave mixing gain, time constant), and performed computer analysis of the measured data.

Dr. Alexander Shumelyuk performed photorefractive characterization in the visible light (orientational, angular and polarization dependences of beam coupling gain factor) and experiments with the transient gain enhancement via periodic phase modulation of the signal wave.

Dr. Konstantin Shcherbin performed photorefractive characterization in the near IR light and the experiments with light induced sensibilization of $Sn_2P_2S_6$ to infrared recording. He was also involved in measurements of orientation dependences in the visible, together with Dr. A. Shumelyuk.

Dr. Serguey Odoulov' contribution to project consists in analysis of possible influence of nondiagonal components of Pockels tensor on the coupling of counterpropagating waves, in processing of angular and polarization dependences of gain factor, in study and analysis of light induced scattering patterns, in writing reports and journal publications.

Description of travels.

No travels were funded by this STCU Project.

Alexander Grabar and Serguey Odoulov attended the Forth International Photorefractive Workshop (Marriott Suites Sand Key, Clearwater Beach, Florida 23-25 August 2006), held by Air Force Research Laboratory, Wright-Patterson, Ohio (AFRL / MLPJ). This trip, supported by European Office of Aerospace Research and Development (EOARD), allowed to present the talks directly related to the progress in present Project, to meet and to discuss the results with the US Partner, Dr. Dean Evans and his colleagues.

Information about major equipment and materials acquired, other direct costs, related to the project.

<u>To ensure successful work on the project the following planned purchases have been made:</u>

1. LASER operating at 671nm, price **1350$**, for the Institute of Physics in Kiev.
2. Computer with Monitor and Printer 6943,5UAH (**1374,95$**) for the Institute of Physics in Kiev.
3. Pure chemicals and materials (tin, sulfur, phosphorus, quartz tubes) 7575UAH (**1500$**), for the Institute of Solid State Physics and Chemistry of Uzhgorod National University (city of Uzhgorod).
4. One He-Ne Laser purchased and the other Laser ЛГН-113 revitalized for total amount 7908 UAH (**1565,94$**), for the Institute of Solid State Physics and Chemistry of Uzhgorod National University (city of Uzhgorod).
5. OSA journal subscription (Optics Letters, J. of the Optical Society of America, B) , (**505$**) .

After purchase of the laser and paying for subscription of scientific journals of the Optical Society of America we still had some money left over that we used as follows:

1. Purchase of Nikon D50 digital photo camera (**830$**)
2. Purchase of office supplies (printer toner and cartridges, office paper, etc.) (**405$**)
3. Upgrade the existing PC (HDD, DVD-RW, RAM, videocapture), (**510$**)

Table Redirection

Reference documents &date (1)	New requested category, or old category with new cost (2)	Requested cost (new) (3)	Original (old) category (4)	Estimated cost (old) (5)	Redirected cost (6) old – new
Quarter <02>					
	Office supplies				405
	Nikon D50 Kit				830
	PS-upgrade (HDD, DVD-RW, RAM, videocapture)				510
Total by L02					1745$

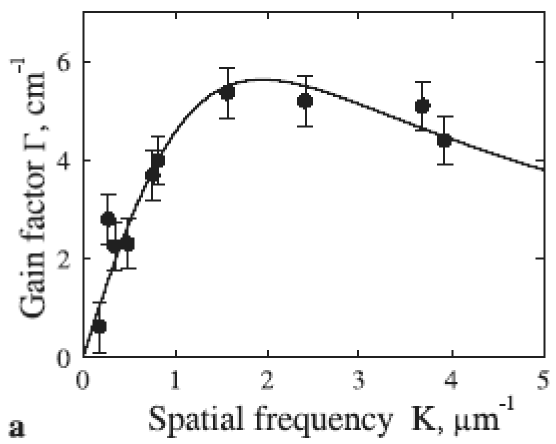
TO-1 The photorefractive characterization of the available and new grown $Sn_2P_2S_6$ samples

Milestone

The Debye screening length of the available $Sn_2P_2S_6$ crystals samples will be determined. (first quarter)

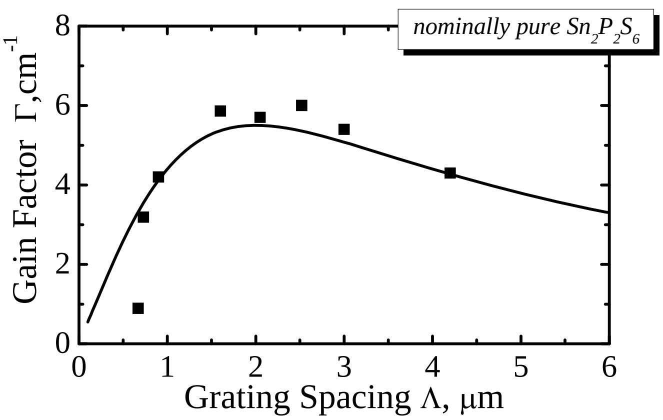
The measurements were started with three available samples, two nominally undoped and the third one deliberately doped with iron. Several typical fringe-spacing dependences (spatial frequency dependences) of beam coupling gain factor for K || OX are shown. They all demonstrate that even the reduced gain factor for the reflection grating geometry is always inferior to 1 cm^{-1}. Also we show the data extracted from measurements of the dynamics at different spatial frequencies. From fitting these data to calculated dependences it is possible to evaluate two characteristic lengths, the diffusion length and Debye screening length.

Both technique confirm that the Debye screening length $\ell_s = 2\pi\sqrt{\frac{\varepsilon\varepsilon_0 k_B T}{e\,N_A}}$ measured with visible light opf moderate intensity falls within the range 1.5 – 3.0 μm for nominally undoped samples.



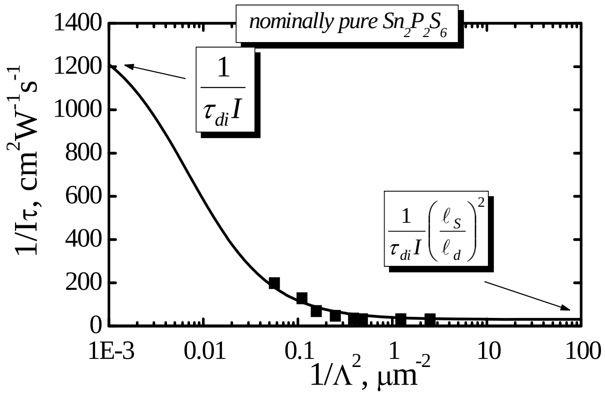
First dependence represents gain factor versus spatial frequency K = 2π/Λ for nominally undoped sample # K1. The screening length estimated from this dependence is quite high, $\ell_s \approx 3\,\mu m$. Solid line shows the best fit of the calculated dependence to the expermental data.

Grating - spacing dependence of the gain factor

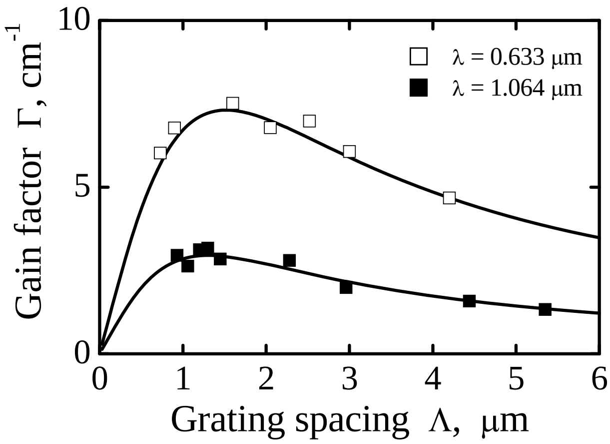


The other example for undoped material shows the screening length about $\ell_s \approx 2\,\mu m$. Solid line shows the best fit of the calculated dependence to the experimental data.

Evaluation of the diffusion length

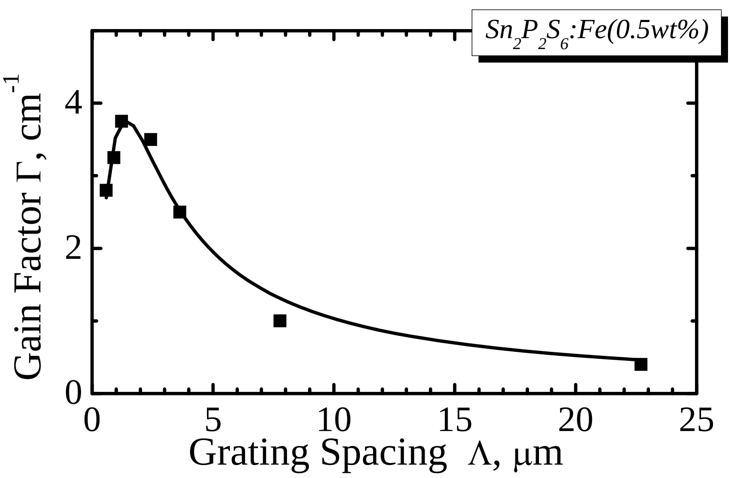


For the same nominally undoped $Sn_2P_2S_6$ sample the estimate for screening length is extracted from the measured dependence of the normalized reciprocal relaxation time versus spatial frequency of the recorded grating. Solid line shows best fit of calculated dependence to the experimental data. The estimate for screening length is the same as that from grating spacing dependence of the gain factor, $\ell_s \approx 2\,\mu m$.



The dependence of gain factor versus grating spacing for two different wavelength (He-Ne and Nd:YAG laser light) is shown for the sample K3. This sample is rather exceptional as it shows smaller screening length compared to other nominally undoped crystals, especially in the near infra red.

Grating - spacing dependence of the gain factor



The iron doped sample was available and similar measurements were performed. It shows the Debye screening length 1.2 μm in the visible, slightly smaller as compared to nominally undoped material but it appeared to be low sensitive in the Near Infrared, at 1.06 μm.

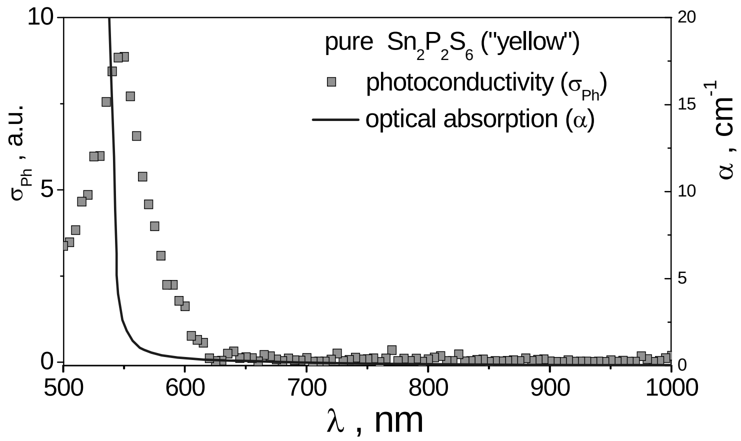
TO-2 ***To grow, to cut and optically finish new nominally undoped $Sn_2P_2S_6$ samples, to perform their optical characterization***

<u>Milestone</u>

Two optically characterized $Sn_2P_2S_6$ crystal samples will be delivered to Kiev group. (second quarter)

According to the workplan two new nominally undoped samples were grown (see one as-grown crystal in the picture above), the samples were cut, poled, polished and repoled once more time.

The optical absorption spectra and photo-conductivity spectra have been measured, both similar to that already known for the undoped material [A. Grabar, Yu. Vysochansky, A. Shumelyuk, M. Jazbinek, G. Montemezzani, P. Guenter, Springer series in Optical Science, vol. 114, pp. **327-362**, 2007].



For these new samples a complete photorefractive characterization has been done also. The largest gain factor **6** cm^{-1} was measured at 630 nm after pre-illumination to the visible light, the Debye screening length being about 2 μm. The particularity of these new samples is in much less developed compensating (secondary) grating as compared to crystals grown previously in the same laboratory in Uzhgorod.

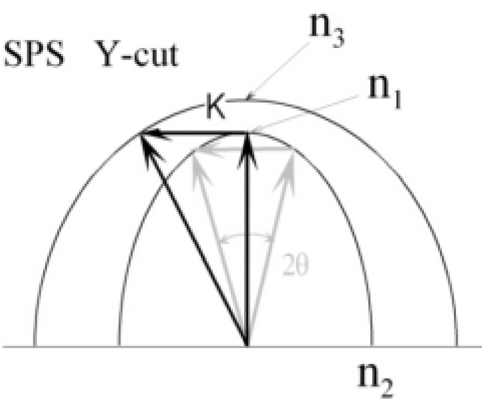
With the mentioned Debye screening length there was a little hope to get an efficient beam coupling in reflection grating geometry. This was confirmed by direct measurement that gave $\Gamma \approx 0.9\ cm^{-1}$.

TO-3 To evaluate experimentally the nondiagonal components of the Pockels tensor

Milestone

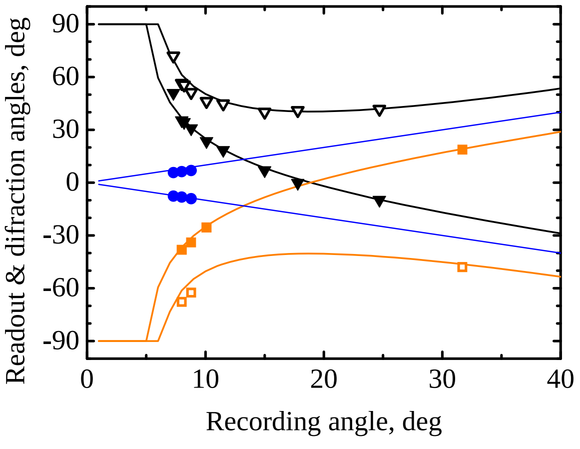
Calculation of the Bragg angles for anisotropic diffraction and evaluation of the electro-optic tensor components. The Bragg angles for anisotropic diffraction based on nondiagonal components of the electrooptic tensor will be calculated. The angular dependences of anisotropic diffraction will be measured and components of the electro-optic tensor will be evaluated from the fit of calculated dependences to experimental data. (third quarter)

From the measured diffraction efficiency and gain factor the values for several previously unknown components of electrooptic tensor have been evaluated. The conclusions from all this efforts is that no one from newly evaluated components of Pockels tensor is larger or even comparable with the largest known component r_{111}. The particular estimated values are r_{333} = 17 pm/V, r_{113} = 20 pm/V, r_{223} = 9 pm/V, assuming r_{111} = 174 pm/V:



Two nondiagonal Pockels tensor components were revealed from the experiments on anisotropic diffraction of isotropically recorded grating. An Y-cut sample of $Sn_2P_2S_6$ was used, with the grating vector of the recorded grating aligned along OX or OZ axis. The anisotropic diffraction was observed in both these geometries at the appropriately chosen angles, extracted from the Evald diagram like shown in figure below.

Two identically polarized waves with the wavevectors shown by gray arrows record a grating with grating vector K. The diffraction with the change of polarization to the orthogonal (anisotropic diffraction) is possible from this grating; the wavevectors of the readout wave and anisotropically diffracted wave are shown by black arrows. Note that for any new angle 2θ between recording waves new position of the readout beam is required to observe the anisotropic diffraction.



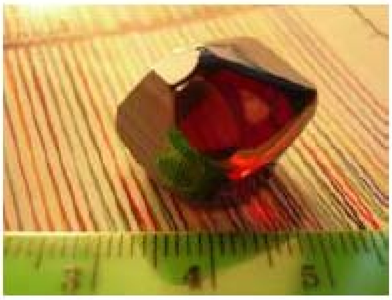
Next Figure shows the dependences of the readout and diffraction angles on the recording angle θ for symmetric incidence of two recording waves to the input face. All values for angles are given in air. The solid lines represent calculated dependences. In trivial case of isotropic readout (blue color), the readout angles are equal to the recording angles so that the dependence is linear. For anisotropic diffraction two pairs of angles meet the Bragg condition, symmetric with respect to crystallographic axis OY. Relevant dependences are shown with black color and brown color, respectively. The observed change in polarization of the diffracted wave and perfect agreement of measured and calculated angular dependences supports the attribution of this type of diffraction to anisotropic diffraction. and allows for evaluation of The particular Pockels tensor components, responsible for this type of diffraction were evaluated (r_{131}/r_{111} = - 0.13 and r_{313}/r_{333} = -0.63). Taking a standard value for r_{111} = 170 pm/V we get the following estimates for r_{131} = -25 pm/V and r_{333} = 22 pm/V.

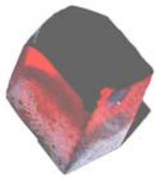
With the present-day level of the signal-to-noise ratio we were unable to find any manifestation of two remaining unknown Pockels components, r_{322} and r_{122}. This conclusion is supported indirectly by the fact that no kind of polarization anisotropic parametric scattering was detected in SPS samples while polarization isotropic scattering was quite pronounced.

T0-4 To grow, cut and opically finish new $Sn_2P_2S_6$ samples with deliberately introduced impurity

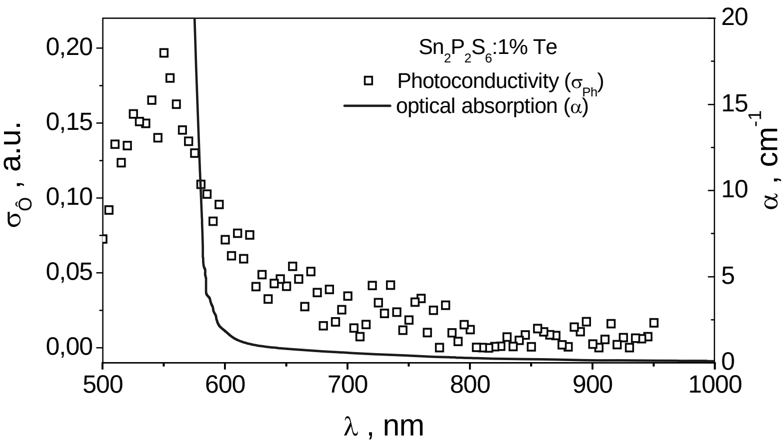
Milestone

Two optically characterized $Sn_2P_2S_6$ crystal samples with 2-times reduced Debye screening length will be synthesized and delivered to Kiev group. (fourth quarter)

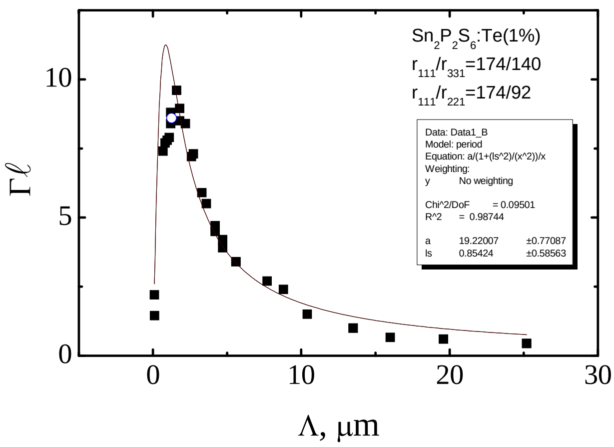




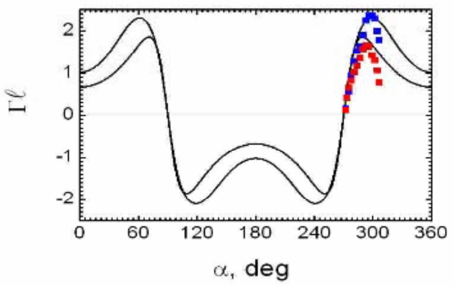
Among different dopants introduced to $Sn_2P_2S_6$ samples during the growth with one of the best appeared to be Tellurium. Typical as-grown sample is shown in this picture (left). From this ingot a sample measuring 9x12x7 mm^3 was cut along the crystallographic directions XxYxZ respectively (right).

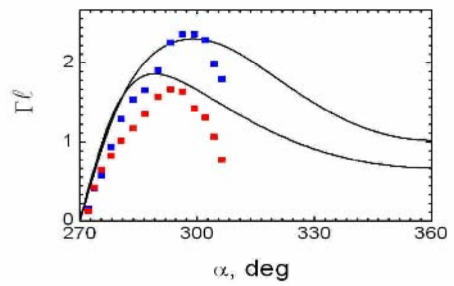


Standard procedure of optical characterization (absorption, photoconductivity) was performed for samples with different Te content. The conclusion is that the optimum density of doping is about 1%; for larger densities of Te the absorption increases too much, so that the net gain because of beam coupling cannot be achieved.



Photorefractive characterization revealed nearly 10 cm^{-1} gain factor for transmission geometry and allowed to make estimate for Debye screening length, $\ell_s = 0.85$ μm. This value is roughly two times smaller than that typical for nominally undoped $Sn_2P_2S_6$. In such a way the expected result for the fourth milestone is achieved by purely technological means (by doping).





With Te-dopped material the reflection gratings have been recorded and studied. In traditional geometry, with the grating vector aligned along OX axis the beam coupling gain factor larger than 1 cm^{-1} was measured, what was expected from the improved Debye screening length.

The grating-vector orientation dependences of the coupling strength were measured for light beams incident to polished Y-faces of the sample
for two eigenpolarizations, shown by blue and red squares. Taking into account that the sample thickness is 12 mm along Y-direction we conclude that the gain factor $\Gamma = 2$ cm^{-1} has been achieved.

To conclude, we demonstrated that by optimizing the direction of beam propagation and polarization of interacting counterpropagating waves in $Sn_2P_2S_6$ crystals with specially selected impurity content it is possible to enhance the exponential gain factor of two beam coupling gain at least two times. Thus the main goal of the project is achieved.

When investigating the beam coupling in nominally undoped $Sn_2P_2S_6$ crystals we developed a new technique that allows for considerable enhancement of the transient beam coupling, via periodic zero-PI phase modulation of one of two interacting waves. This technique is described, together with the first experimental confirmation in article accepted for publication in Optics Letters:

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Transient gain enhancement in photorefractive crystals with two types of movable charge carrier

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